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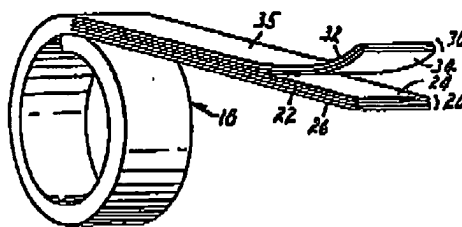
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(64) Linerless double-coated pressure-sensitive adhesive tape.

(67) A linerless double-coated pressure-sensitive adhesive tape of the prior art is wound directly upon itself into a roll that can later be unwound without delamination or offsetting of adhesive. The novel tape differs from the prior tape in that the pressure-sensitive adhesive at each of its faces is substantially solvent-free, crosslinked alkyl acrylate polymer and can be aggressively tacky for uses requiring high-performance adhesion.

Also provided is a composite of two pressure-sensitive adhesive tapes which are releasably adhered together adhesive face to adhesive face and then put to individual uses. The tapes are separable because their facing adhesives are substantially solvent-free, crosslinked alkyl acrylate polymers which can be aggressively tacky for uses requiring high-performance adhesion.

**FIG. 2**
**EP 0 121 430 A2**

Croydon Printing Company Ltd.

**0121430****LINERLESS DOUBLE-COATED PRESSURE-SENSITIVE ADHESIVE TAPE****Field of the Invention**

5 The invention concerns a linerless double-coated pressure-sensitive adhesive tape which can be wound directly upon itself into a roll for storage and shipment. The tape comprises a flexible support, at each face of which is a pressure-sensitive adhesive. The invention also concerns a composite having a first tape with a pressure-sensitive adhesive coating on both faces and a second tape with a pressure-sensitive adhesive coating on at least one face, the first and second tapes being releasably adhered together adhesive face to adhesive face.

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**Background Art**

15 Almost all double-coated pressure-sensitive adhesive tapes are wound up with disposable, low-adhesion liners which are discarded when the tapes are unwound for use. Many double-coated tape dispensers are equipped to wind up the liner as the tape is dispensed. This not only makes the dispenser more expensive and awkward to handle, but the user must occasionally take the time to discard the wound liner. Disposal of the liner is especially troublesome in robotic systems.

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25 U.S. Patent No. 2,889,038 (Kalleberg) discloses a linerless double-coated pressure-sensitive adhesive tape wound upon itself in roll form and comprising a flexible support having on opposite faces chemically different and physically incompatible pressure-sensitive adhesive layers. Although the Kalleberg patent calls both layers "aggressively tacky", the patented linerless transfer tape is not currently used when high performance is required. Even though Kalleberg tapes are currently on the market, it is believed that at the present time all double-coated pressure-sensitive adhesive tapes which have high performance are wound up with disposable, low-adhesion liners. Furthermore, the adhesive layers of the Kalleberg

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tapes being marketed have a mottled appearance, and it usually is necessary to limit the thickness of each layer to about 0.1 mm.

Disclosure of Invention

5           The invention concerns a linerless double-coated pressure-sensitive adhesive tape and a composite which incorporates a linerless double-coated pressure-sensitive adhesive tape, which tape like that of the Kalleberg patent comprises a flexible support, at each face of which is a pressure-sensitive adhesive. The tape can be wound directly upon itself into a roll that can later be unwound without delamination or offsetting of adhesive. The composite includes the linerless double-coated pressure-sensitive adhesive tape as a first tape and a second tape having a flexible support on at least one face of which is a pressure-sensitive adhesive layer, the first and second tapes being releasably adhered together adhesive face to adhesive face. Unlike the Kalleberg tape, the adhesive faces of the novel tape and the composite can have truly high performance and can have adhesive and cohesive values equal to those of any pressure-sensitive adhesive tape now on the market. Also, unlike the Kalleberg tape, the novel tape does not require chemically different and physically incompatible adhesives at the two faces of its flexible support and the composite does not require chemically different and physically incompatible adhesives at the releasably adhered faces of the first and second tape. Instead, unwindability in the novel tape and separability in the composite are attained in that the pressure-sensitive adhesive at each adhesive face

(a) is a polymer of predominantly alkyl acrylate, the alkyl groups of which have an average of four to twelve carbon atoms,

(b) is substantially solvent-free,

(c) is crosslinked, and

(d) affords at each face a 180° Peelback Value

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(as defined below) of at least 10 N/dm.

By "substantially solvent-free" is meant that the pressure-sensitive adhesive contains less than three percent by weight of solvent. The pressure-sensitive adhesive is sufficiently crosslinked when, on attempting to dissolve in heptane at room temperature, the undissolved gel component exceeds 50%.

A pressure-sensitive adhesive which provides a 180° Peelback Value of only 10 N/dm may be characterized as moderately tacky, being just tacky enough to assure reliable adhesion to ordinary paper, metal, glass, plastic, and painted substrates. When the adhesive at each face of the linerless double-coated pressure-sensitive adhesive tape is aggressively tacky, the novel tape can nevertheless be readily unwound from a linerless roll without delamination or offsetting of adhesive, even after months of storage. When the adhesive at each face of the novel composite is aggressively tacky, its first and second tapes can nevertheless be readily peeled apart without delamination or offsetting of adhesive, even after months of storage. Each face of the novel tape can have a 180° Peelback Value as high as that of any pressure-sensitive adhesive tape now on the market, e.g., at least 30 N/dm.

It is surmised that if the adhesive were not substantially solvent-free, the solvent would allow the polymer chains to knit across adjacent convolutions during prolonged storage in roll form, such that perfect separation could no longer be assured. In the present state of the art, it would not be commercially feasible to coat a pressure-sensitive adhesive from solution and obtain a pressure-sensitive adhesive layer which is substantially solvent-free such that aggressively tacky adhesive layers could be reliably separated without delamination or offsetting of adhesive.

To keep the amount of solvent to a minimum, the novel tape and the tapes of the novel composite are preferably made using photopolymerization as in U.S. Patent

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No. 4,181,752 (Martens et al). No solvents are used when doing so. Although there inevitably are impurities in the starting materials which may have a solvent effect, such impurities would not comprise as much as three percent by weight of the pressure-sensitive adhesive.

The preferred procedure for preparing the linerless double-coated pressure-sensitive adhesive tape comprises the steps of

(1) coating onto each face of a flexible support a substantially solvent-free photopolymerizable monomer mixture comprising an alkyl acrylate, the alkyl groups of which have an average of 4-12 carbon atoms, and a crosslinker, and

(2) then exposing each coating to ultraviolet radiation to polymerize the acrylate to provide a pressure-sensitive adhesive layer which

(a) is crosslinked and

(b) affords at each face a 180° Peelback Value of at least 10 N/dm.

The resulting double-coated pressure-sensitive adhesive tape can be wound up into roll form adhesive face to adhesive face and later unwound without delamination or offsetting of adhesive. When the flexible support of the double-coated tape is porous, it may be embedded in a single layer of pressure-sensitive adhesive. Whether or not the flexible support is porous, there may be a pressure-sensitive layer on each face of the support and those two layers may either be identical or different from each other.

The preferred procedure for making the novel composite comprises the steps of

(1) coating onto each face of a first flexible support a substantially solvent-free photopolymerizable monomer mixture,

(2) coating onto one face of a second flexible support a substantially solvent-free photopolymerizable mixture,

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each said photopolymerizable mixture comprising an alkyl acrylate, the alkyl groups of which have an average of 4-12 carbon atoms, and a crosslinker, and

(3) exposing each coating to ultraviolet radiation to polymerize the acrylate to provide a pressure-sensitive adhesive layer which

(a) is crosslinked and

(b) affords at each face a 180° Peelback Value of at least 10 N/dm.

(4) superimposing the resulting two tapes adhesive face to adhesive face, thus providing the novel composite. Those two tapes can later be peeled apart without delamination or offsetting of adhesive.

When the first flexible support of the novel composite is porous, it may be embedded in a single layer of pressure-sensitive adhesive. Whether or not that flexible support is porous, there may be a pressure-sensitive layer on each face of the support and those two layers may either be identical or different from each other.

Preferably prior to the aforementioned step (1) and before adding the crosslinker, the photopolymerizable mixture is first partially polymerized by ultraviolet radiation to provide a syrup having a coatable viscosity, e.g., 300 to 20,000 centipoises. After adding the crosslinker, the syrup is coated out and then exposed to ultraviolet radiation in an inert environment to complete the polymerization while simultaneously crosslinking the polymer. An inert environment may be provided by plastic film overlays which are fairly transparent to ultraviolet radiation and have low-adhesion surfaces. Biaxially-oriented polyethylene terephthalate film which is about 75% transparent to ultraviolet radiation of 320 to 370 nm is very useful. If instead of covering the polymerizable coating, the polymerization is to be carried out in an inert atmosphere, the permissible oxygen content of the

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inert atmosphere can be increased by mixing into the polymerizable composition an oxidizable tin compound as taught in U.S. Patent No. 4,303,485 (Levens), which also teaches that by doing so, thick coatings can be polymerized in air. If the monomer mixture is partially polymerized and the polymerization is to be completed in situ by heat, it is usually necessary to add additional heat-activatable polymerization initiator to the partially polymerized composition.

Solvents can also be substantially avoided by in situ polymerization by electron beam as suggested in U.S. Patent No. 4,243,500 (Glennon). See also U.S. Patent No. 2,956,904 (Hendricks).

The pressure-sensitive adhesive of the novel tape can be substantially solvent-free if polymerized and coated from aqueous systems. However, coatable aqueous emulsions usually contain organic materials which are themselves solvents for pressure-sensitive adhesives, and it may be difficult to eliminate them substantially.

To attain high 180° Peelback Value, the pressure-sensitive adhesive of the novel tape may be made using one or more copolymerizable monomers having strongly polar groups such as acrylic acid, methacrylic acid, itaconic acid, acrylamide, methacrylamide, acrylonitrile, methacrylonitrile, or N-vinyl-2-pyrrolidone. Amounts of such copolymerizable monomers up to about 12 mol percent (25 mol percent when using the pyrrolidone) of total monomer tend to provide increased 180° Peelback Value while higher amounts tend to result in decreased 180° Peelback Values unless the tape is heated when the bond is formed. Where heat is to be applied in forming bonds, a strongly polar copolymerizable monomer can provide nearly 50 mol percent of total monomer.

Other useful copolymerizable monomers include vinyl toluene, vinyl chloride, vinylidene chloride, styrene, and diallyl phthalate. One or more of these may be employed in amounts up to 5 mol percent of total monomer

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without appreciably changing any of the properties of the novel tape.

At constant levels of strongly polar and other copolymerizable monomer, the highest 180° Peelback Values have been attained when the alkyl acrylate monomer includes both a C<sub>5-14</sub> acrylate and a C<sub>1-4</sub> acrylate. Preferred as the C<sub>5-14</sub> acrylate is isooctyl acrylate obtained by esterifying acrylic acid with isooctyl alcohol, a mixture of various isomers of octyl alcohol which is readily available commercially at relatively low prices. Preferred C<sub>1-4</sub> acrylates are obtained from methyl, ethyl or n-butyl alcohol, which also are readily available at reasonable prices. Acrylic acid is a preferred strongly polar copolymerizable monomer, being both low in cost and easily copolymerizable with the alkyl acrylate, especially by the procedure of the aforementioned U.S. Patent No. 4,181,752.

Homopolymers of useful alkyl acrylates are weak internally but when crosslinked should develop useful internal strengths as indicated by Shear Values of at least 3 minutes. Higher internal strengths can be achieved through the use of one or more of the strongly polar copolymerizable monomers mentioned above.

Among crosslinking agents useful in making the novel tape by photopolymerization as in the above-cited Martens patent are those which also function as photopolymerization initiators such as 2,4-bis(trichloromethyl)-6-p-methoxystyryl-s-triazine. The chromophore-substituted-halomethyl-s-triazines of U.S. Patents No. 4,329,384 and 4,330,590 are especially useful where a high 180° Peelback Value is desired. Other useful photocrosslinkers are polyacrylic-functional monomers such as trimethylolpropane triacrylate; pentaerythritol tetraacrylate; 1,2-ethylene glycol diacrylate; 1,6-hexanediol diacrylate; and 1,12-dodecanediol diacrylate. Each of these photocrosslinkers is preferably used within the approximate range of 0.05 to 0.5 percent by weight of the polymerizable monomers.



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While the reason for the surprising linerless capability of the novel double-coated tape and the surprising capability of separating the tapes of the novel composite are unknown, it is surmised that the crosslinking  
5 knits the polymer chains together at each face of the tape such that the contacting pressure-sensitive adhesive surfaces separate cleanly at their interface when peeled apart. Other phenomena may also be involved. For  
10 example, when the polymer is made using one or more copolymerizable monomers having strongly polar groups, those groups may turn inwardly, leaving the relatively nonpolar alkyl chains at the surface, thus providing a release effect.

To enhance immediate adhesion to relatively rough  
15 or uneven surfaces, the flexible supports of the novel tape and the novel composite may be a resilient foam such as the backing foam of Canadian Patent No. 747,341 (Engdahl et al). Another technique for enhancing immediate adhesion to relatively rough or uneven surfaces is to incorporate glass  
20 microbubbles into the pressure-sensitive adhesive as taught in U.S. Patent No. 4,223,067 (Levens). Other materials which can be blended with the polymerizable mixtures include reinforcing fibers and agents, fire retardants, and inert fillers. If the polymerization is to be effected by  
25 ultraviolet radiation, such materials should allow the radiation to penetrate into the entire coating during the photopolymerization step.

When the flexible support of the novel tape or either flexible support of the composite tape is a  
30 transparent plastic film such as polyvinyl chloride, that tape can be almost perfectly transparent. Such a tape is especially useful for laminating transparent panels, for applying labels having transparent areas, and for applying transparent protective coverings. The composite may be put  
35 to use such as are described in connection with Fig. 4 of the drawings.

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180° Peelback Value

At about 22°C, tape is adhered by its adhesive to a glass or stainless steel test plate under the weight of a 4.5 kg hard rubber roller, 2 passes in each direction. The free end of the tape is attached to a scale, and the test plate is moved away from the scale at an angle of 180° and a rate of about 3.8 cm per second.

Shear Value

At about 22°C, tape is adhered by its pressure-sensitive adhesive to a stainless steel plate under the weight of a 4.5-kg hard rubber roller, 2 passes in each direction, with a free end of tape extending beyond the plate and the adhesive contact area being one-half by one-half inch (1.27 by 1.27 cm). After thirty minutes, the plate is positioned 2° from the vertical to prevent peeling, and a 1-kg mass is suspended from the free end. The time at which the mass falls is noted and the test is discontinued if the tape has not failed after 10,000 minutes.

The Drawings

In the drawings:

Figure 1 is a schematic isometric view of a linerless double-coated pressure-sensitive adhesive tape of the invention wound upon itself in roll form.

Figure 2 schematically illustrates a composite of two pressure-sensitive adhesive tapes wound up together into a single roll, one of those tapes being a linerless double-coated pressure-sensitive adhesive tape of the invention.

Figure 3 illustrates a use for the composite of Figure 1.

Figure 4 illustrates a use to which another composite of the invention may be put.

The linerless double-coated tape 10 of Figure 1 has a porous flexible cloth support 12 embedded in a single

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has a flexible film support 52 and two pressure-sensitive adhesive layers 54, 56. Adhered by the upper adhesive layer 56 is a thin flexible article 58 which can be peeled away, carrying with it the second tape 50 to be remounted by its adhesive layer 54.

In the following examples, all parts are given by weight.

#### Example 1

To 100 parts of isooctyl acrylate was added 0.04 part of 2,2-dimethoxy-2-phenyl acetophenone photoinitiator (obtainable as "Irgacure" 651). This was partially polymerized by exposure to ultraviolet radiation to provide a viscosity of about 3000 cps. To this was added 0.2 part of 1,6-hexanediol diacrylate crosslinker and an additional 0.1 part of the photoinitiator to provide a first syrup.

A mixture of 96 parts of isooctylacrylate, 4 parts of acrylic acid, and 0.04 part of photoinitiator ("Irgacure" 651) was partially polymerized by exposure to ultraviolet radiation to provide a coatable viscosity of about 3000 cps. After mixing in 0.2 part of 1,6-hexanediol diacrylate and an additional 0.1 part of the photoinitiator to provide a second syrup, this was poured onto the low-adhesion surface of a first disposable plastic film. A film of unprimed, unplasticized, unpigmented polyvinyl chloride of 0.1 mm thickness was laid over the poured-out second syrup, and the composite was drawn beneath a first knife to squeeze the second syrup to a uniform thickness of about 0.05 mm. The first syrup was poured onto the exposed face of the vinyl film, and to this was joined, at a second knife, the low-adhesion surface of a second disposable plastic film. The spacing beneath the second knife was

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